MEASUREMENT OF CHARGE TRANSFER RATE COEFFICIENT BETWEEN GROUND-STATE N²⁺ ION AND He AT ELECTRON-VOLT ENERGIES

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ABSTRACT

The charge transfer rate coefficient for the reaction $N^{2+}(2p\ ^2P^o) + He \to$ products is measured by recording the time dependence of the N^{2+} ions stored in an ion trap. A cylindrical radio-frequency ion trap was used to store N^{2+} ions produced by laser ablation of a solid titanium nitride target. The decay of the ion signals was analyzed by single exponential least-squares fits to the data. The measured rate coefficient is $8.67(0.76) \times 10^{-11}$ cm³ s⁻¹. The N^{2+} ions were at a mean energy of 2.7 eV while He gas was at room temperature, corresponding to an equivalent temperature of 3.9×10^3 K. The measured value is in good agreement with a recent calculation.

Subject headings: atomic data — atomic processes — methods: laboratory

1. INTRODUCTION

In the interstellar gas, multiply charged ions are formed and co-exist with neutral atoms of hydrogen and/or helium. Since the interstellar gas is not in thermodynamic equilibrium because of its low density (Steigman 1973, 1975), the distribution of elements among its various ionization stages is governed by several completing processes including charge transfer, dielectronic recombination, radiative recombination, electron impact ionization, and photoionization. Depending on the environment, charge transfer can be the dominant process that controls the states of ionization in astrophysical plasmas (Dalgarno 1985; Shields 1990; Dalgarno & Fox 1994). For a realistic modeling of these astrophysical plasmas, it is of paramount importance that the charge transfer rate coefficients be known accurately at the temperatures of these environments (Shull 1993).

In the past, the Landau-Zener model was used to estimate the charge transfer cross sections and rate coefficients. However, this technique may fail at low energies. Vigorous quantal calculations have been used to provide more accurate results. At present, there are few experimental tests of these calculated values and their accuracy. Our recent measurement of the charge transfer rate coefficient of O^{2+} and He (Kwong & Fang 1993; Fang & Kwong 1994) indicates that the measured rate coefficient is three orders of magnitude smaller than the full quantal calculations by Gargaud, Bacchus-Montabonel, & McCarroll (1993) and an order of magnitude smaller than the Landau-Zener calculation of Butler, Heil, & Dalgarno (1980).

Charge transfer of N^{2+} and He at eV energies can play an important role in stellar chemistry as a source of N^+ ions in nebulae (Dalgarno 1985; Sun et al. 1996). Three calculations have been made at this energy range. Calculations made by Nikitin & Reznikov (1992) and Lafyatis, Kirby, & Dalgarno (1993) were based on the Landau-Zener model while Sun et al. (1996) carried out a sophisticated multistate close-coupling quantal calculation. However, all these calculations disagree with the measurement of Sadilek et al. (1990) using beam scattering at eV energies. In this paper, we report a measurement of the total charge transfer rate coefficient of the reaction by using a laser ablation ion source and ion storage technique.

2. EXPERIMENTAL METHOD

The measurement of the ground-state thermal energy charge transfer rate coefficient between N^{2+} ion and helium is carried out by using the technique of laser-ablation ion source and ion storage that is uniquely developed in this laboratory and dedicated for low-energy charge transfer studies. We have described in detail this measurement technique in our previous publications (Kwong et al. 1990; Fang & Kwong 1994, 1995).

The N^{2+} ions are produced by laser ablation of solid titanium nitride (TiN) targets. Titanium nitride was chosen as the target material because of the distinct mass-to-charge ratio difference between N^{2+} ions ($m/q = 7$) and other low-charge-state ions produced from the target (N^+, $m/q = 14$; Ti^+, $m/q = 48$; Ti^{2+}, $m/q = 24$; Ti^{3+}, $m/q = 16$; etc.). The measurement of Ti^{q+} + He will be reported elsewhere. We used the output of the second harmonic (532 nm) of a pulsed Nd:YAG laser as an ablation source. In this series of measurements, approximately 1 mJ of the second harmonic of the Nd:YAG laser was used. The power density was estimated to be about 10^8 W cm⁻² with laser pulse duration of about 25 ns. The low power density of the ablation source eliminates the formation of higher charge states of titanium and nitrogen. The prominent feature of this method is that no source gas is used in ion production, therefore the complexities and uncertainties caused by the interaction between the ions and the source gas are totally eliminated. A cylindrical radio-frequency (rf) quadrupole ion trap serves as a mass-selective ion storage device. The trapping parameters (rf f = 1.44 MHz, amplitude V_0 = 350 V, and dc bias U_0 = 29 V) were chosen to selectively store N^{2+} ions. The axial well depth was D_z = 26.6 eV and the radial well depth was D_r = 35.6 eV for N^{2+} ions. The low charge states of titanium ions and singly charged nitrogen ions were excluded from the trap. To detect the stored ions, two extraction pulses (+190 V and −200 V) were applied simultaneously to the upper and lower end caps of the trap, respectively. These push and pull extraction pulses empty all the stored ions into a 0.3 m time-of-flight (TOF) mass spectrometer where the signal was detected by a 1 inch (2.5 cm) diam dual microchannel plate. The TOF mass spectrum was recorded by a transient digitizer and stored in a
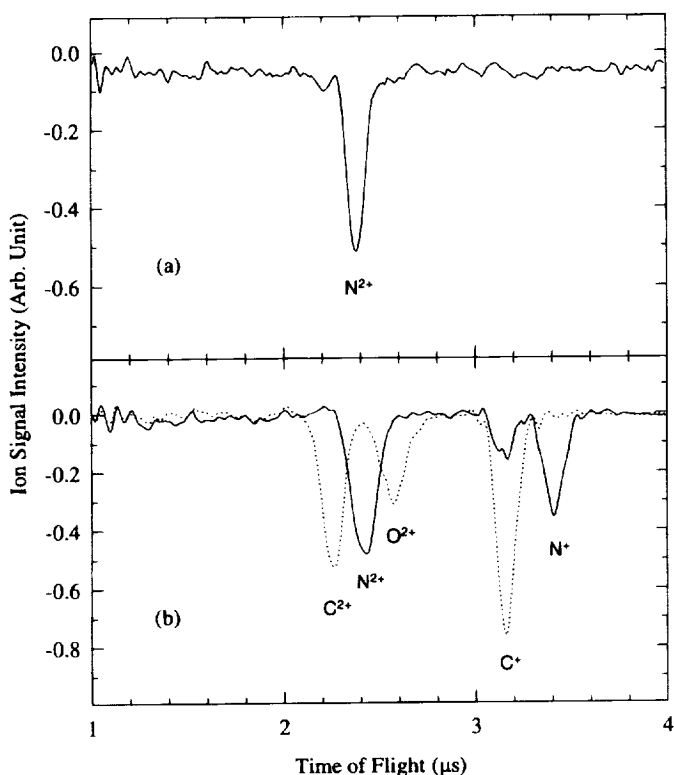
FIG. 1.—(a) TOF mass spectrum of the laser-produced N^{2+} ions which have been stored in the trap. The parameters of the trap were set to exclude the ions with $m/q \geq 14$. (b) TOF mass spectra of the stored ions produced by electron impact dissociative ionization of N_2 (solid line) and CO (dotted line) gases. The parameters of the trap were set to store ions with their m/q up to 14.

computer for later analysis. The TOF signal was used to identify the ion species while the signal intensity served to measure the population of the ions stored in the trap prior to their extraction. Figure 1a is a typical TOF mass spectrum of the laser-produced ions stored in the trap under above-mentioned trapping condition. Only N^{2+} ions are stored in the trap. For the purpose of comparison, Figure 1b shows the TOF mass spectra of stored N^{2+}, N^+, C^{2+}, O^{2+}, and C^+ ions, produced by electron impact on ultrahigh-purity N_2 and CO gases. The number of N^{2+} ions stored in the trap was about 10^3. The storage time ($1/e$) was in excess of 10 s at a base pressure of 4×10^{-10} torr.

The charge transfer rates were obtained by measuring the relative number of N^{2+} ions remaining in the trap as a function of time, in the presence of helium gas of known density. To minimize both the short-term and the long-term ion signal fluctuation and drift caused by the variation of laser power and the changing surface condition of the target when the ablation laser gradually drills into the target surface, the ion signals were measured, alternately, at a delay time t and at the shortest delay time $t_0 = 0.4$ s. Each pair of measurements consisted of 20 laser shots. More than 10 such pairs of measurements were made for each time t. The intensity ratio, $I_r = I_t/I_{t_0}$, is computed to obtain a normalized relative intensity. The storage time, t, is then scanned with a delay time increment, δt, to obtain an ion decay curve.

In this measurement, helium gas with ultrahigh-purity grade was used (Linde, ultrahigh purity, 99.999%). Helium gas was introduced into the vacuum chamber through a gas handling system. The operation of gas handling system to

minimize contamination was described earlier (Fang & Kwong 1995). The purity of the He gas in the vacuum chamber was confirmed by a quadrupole mass analyzer.

3. DATA ANALYSIS AND RESULTS

Figure 2 is a plot of the relative ion signal intensities versus storage time, t, after laser ablation at four different helium gas pressures. The error bar for each data point represents the standard error (1 σ), derived statistically from the measurements resulted by hundreds of laser shots. Each solid line is a weighted least-squares fit to the data by a single exponential decay function. The slope of the ln-linear fit gives the charge transfer rate at a given helium pressure. The uncertainty of the charge transfer rate is determined by the weighted least-squares fit algorithm.

Figure 3 is a plot of the charge transfer rate between N^{2+} ion and He as a function of helium pressures. The slope in the figure is obtained by the weighted least-squares fits to a linear function. The charge transfer rate coefficient for N^{2+} ion and He obtained from slope of Figure 3 is $8.67(0.76) \times 10^{-11}$ cm^3 s^{-1}. The uncertainty presented is the quadratic sum of the uncertainty of the helium density measurements (8%) and the uncertainty due to the statistical fluctuation of the ion intensities derived from the fitting.

4. DISCUSSION AND SUMMARY

4.1. Electronic State of the Stored N^{2+} Ions

N^{2+} ions produced by laser ablation can be in a variety of excited electronic states immediately after they are produced. They rapidly cascade to their ground state and low-lying metastable states through allowed transitions and collisional deexcitations by plasma electrons. We have shown in our previous publications that the ions produced by laser ablation at low laser power densities are in their ground state (Kwong & Fang 1993; Fang & Kwong 1994,
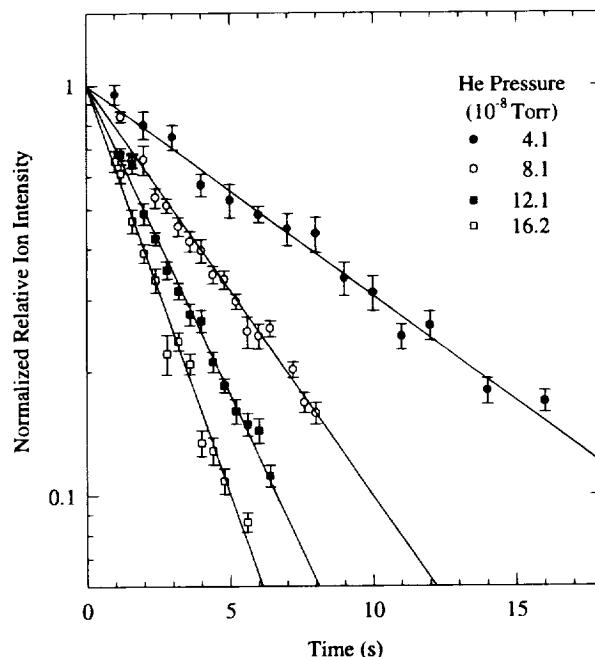

FIG. 2.—The decay curves of normalized relative intensity of N^{2+} ions vs. storage time at four different pressures of He. The uncertainty (1 σ) is due to the statistical fluctuation of the ion signal. Solid lines are the least-squares fits to a single exponential function.
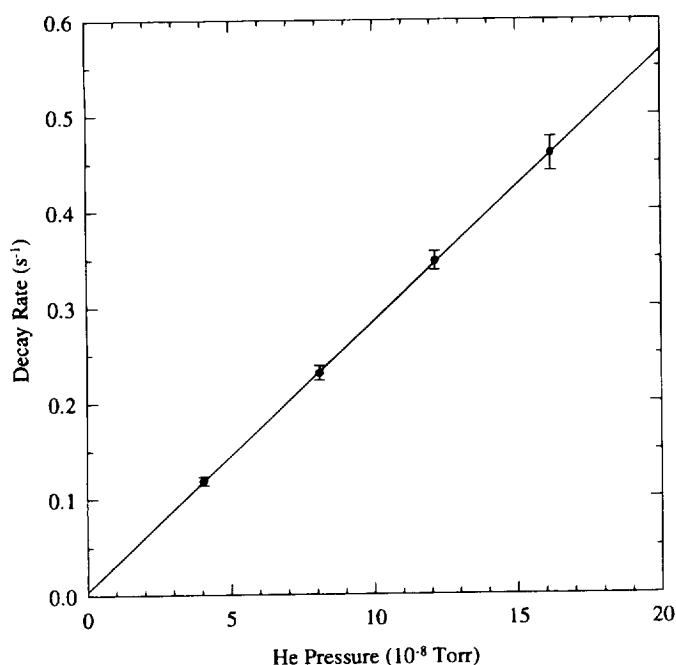
FIG. 3.—N^{2+} ion decay rate vs. He pressure. Each error bar represents the statistical uncertainty of 1 σ. The slope of the straight line fit gives the charge transfer rate coefficient of the N^{2+} ion with He.
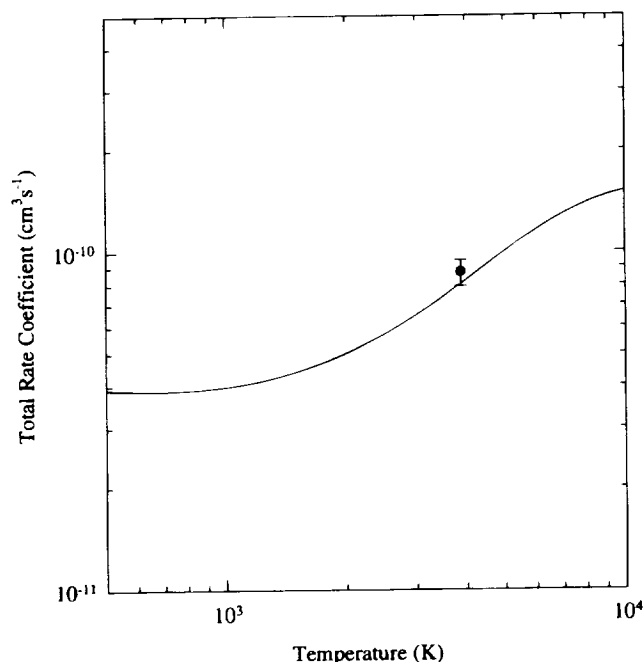

FIG. 4.—Total rate coefficients of charge transfer between N^{2+} ($2p\ ^2P^o$) and He. The filled circle represents the present work. The solid line is the calculated result by Sun et al. (1996).

1995). Furthermore, N^{2+} has only one low-lying metastable state, $2s2p^2(^4P)$, with three fine-structure levels. Their lifetimes are: 0.98 ms for $^4P_{1/2}$, 13.4 ms for $^4P_{3/2}$, and 3.2 ms for $^4P_{5/2}$ (Fang, Kwong, & Parkinson 1993). At the time we start to measure the ion intensity (0.4 s after the laser ablation), the stored metastable N^{2+} ions, if there are any, should have decayed to the $2s^22p(^2P^o)$ ground state.

4.2. Mean Energy and Temperature

The mean energy of the stored N^{2+} ions is about 2.7 eV which is estimated from the potential well depth of the trap (Kwong & Fang 1993; Knight & Prior 1979). This mean energy corresponds to an ion temperature of about 2.1×10^4 K, calculated by using the relation $\bar{E} = 3/2kT$ since the velocity distribution of the ions in a trap is nearly thermal (Knight & Prior 1979). Considering the temperature of the He gas in the experiment is about 300 K (room temperature), it is more appropriate to introduce an equivalent temperature to compare with results where both reactants are in thermal equilibrium (Dickinson 1995). The equivalent temperature, T_{equiv}, is determined by the mean

relative velocity between ion and neutral atom with the ion temperature T_i and the neutral temperature T_n. The relation between these temperatures is given by

$$\frac{T_{equiv}}{\mu} = \frac{T_i}{m_i} + \frac{T_n}{m_n},$$

where m_i and m_n are the mass of ion and the mass of neutral, respectively, and μ is their reduced mass. The equivalent temperature for this measurement is 3.9×10^3 K.

4.3. Summary

Figure 4 illustrates the comparison of our measured charge transfer rate coefficient at temperature 3.9×10^3 K with the calculated total rate coefficient by Sun et al. (1996). The calculated value is within 1 σ of our measurement.

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